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A Method of Determining Dielectric Constants of Liquids

A METHOD OF DETERMINING DIELECTRIC CONSTANTS OF LIQUIDS BY UNDAMPED OSCILLATIONS

BY

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I HEREBY RECOMMEND THAT THE THESIS PREPARED UNDER MY SUPERVISION BY WILLIAM HENRY HYSLOP

ENTITLED A METHOD OF DETERMINING DIELECTRIC CONSTANTS OF LIQUIDS BY UNDAMPED OSCILLATIONS

BE ACCEPTED AS FULFILLING THIS PART OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN PHYSICS

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H I S T O R I C A L

I INTRODUCTION

The first published results on the measurements of dielectric constants were those obtained by Faraday in his classical experiments in which he demonstrated that the capacity of a condenser depends upon the substance used as a dielectric. (Experimental Researches in Electricity, Vol.I, p 394). Finding that the capacity of a condenser in which sulphur was used as a dielectric was 1.66 times as great as that of the same condenser with air as the dielectric, Faraday termed this value the "specific inductive capacity" of sulphur. The term "dielectric constant", however, has been more generally used of late years. Previous to the year 1889, all methods of determining the dielectric constant of a substance made use of electric fields set up by either direct or slowly alternating currents. The discoveries of Hertz in 1888 suggested new methods in which high frequency currents were employed. The first of these methods, that of J.J. Thomson, was developed in 1889, and employed a frequency of approximately twenty five millions. All high frequency methods may be divided into three general classes: (1) those based upon Maxwell's electromagnetic theory of light; (2) capacity methods; and (3) those based upon the theory of dielectric polarization.

In the development of the electromagnetic theory of light, Maxwell showed that the velocity of propagation of an electromagnetic disturbance is expressed by the relation $v = 1/\sqrt{k\mu}$, where k is the dielectric constant of the medium and μ is the magnetic permeability of the medium. (Treatise on Electricity and Magnetism, Vol.II, Sec. 785). For all dielectric media, μ is taken as unity. From the ordinary optical definition, the index of refraction of a medium,

$n = \frac{V_o}{V}$, where V_o is the velocity of propagation in free ether and V the velocity in the dielectric medium. Considering the value of the dielectric constant of free ether as unity, it follows that $k = n^2$. Methods of the first class, then, involve the determination of n, from which k may be calculated.

Capacity methods are based upon Faraday's definition of the dielectric constant, expressed by the relation $k = \frac{C_s}{C_a}$, where C_s represents the capacity of a condenser with the given substance as the dielectric, and C_a the capacity of the same condenser with air as the dielectric. Methods in this class employ circuits of low resistance in which the frequency of the currents depends upon the inductance and capacity of the circuit. The frequency f is expressed by the relation deduced by Lord Kelvin, $f = 1/2\pi\sqrt{1/LC - R^2/4L^2}$. (Phil. Mag. Vol.5, Ser.4, p 393, 1853). Where R is so small that the second term under the radical may be neglected, the expression reduces to the form $T = 2\pi\sqrt{LC}$, since the period T is the reciprocal of the frequency. With two exceptions, the capacity methods with high frequencies have been resonance methods. In a resonance method two oscillatory circuits are usually employed. (Fig.1). The primary circuit consists of an inductance L_1 and a capacity C_1 in series with a spark-gap. Energy is supplied by an induction coil or some other source of high potential. When sparks are passing across the gap, electrical oscillations take place in the circuit, the period of these oscillations being determined by the values L_1 and C_1. The secondary circuit consists of an inductance L_2, a capacity C_2, and some kind of indicating device A for determining whether the current in the secondary circuit is a maximum. If the two circuits are coupled either by capacity or inductance, or directly as in some

cases, oscillations in the primary induce oscillations in the secondary. When adjustments are made until the products L_1C_1 and L_2C_2 are equal, the two circuits have equal periods and are said to be in resonance, a condition which is detected by a maximum reading of A. By making use of two secondary circuits, "null" methods have been devised, in which the current effects in the two secondary circuits balance each other and the indicating device shows a zero deflection at resonance.

The third class of methods of determining the dielectric constant has been used but twice for high frequencies. These methods are based upon the Poisson-Mossotti theory of dielectric polarization in which the relation $k = 1 + 4\pi\chi$ is deduced, where χ may be termed the "dielectric susceptibility" of the substance. Graetz and Fomm suspended a small thin-walled ellipsoid of revolution, made from hard rubber and filled with water, between the plates of an air condenser which formed part of an oscillating circuit. (Wied. Ann. 54, p 626, 1895). The ellipsoid was placed with its major axis at an angle of about 45° with the direction of the field and the deflection produced by the rapidly alternating field was measured. The value obtained for k was 73.54 and was independent of the frequency, which varied from 10^6 down to 100. Later, Beaulard made determinations of the dielectric constant of water and of alcohol by using quartz ellipsoids filled with the liquid and placed in a field having a frequency 6×10^6. (Jour. de Phys. 5, p 165, 1906). His values were much lower than those obtained by other observers.

II WAVE METHODS

1. Refraction Methods. In refraction methods the substance to be tested is placed in a vessel having the form of a triangular

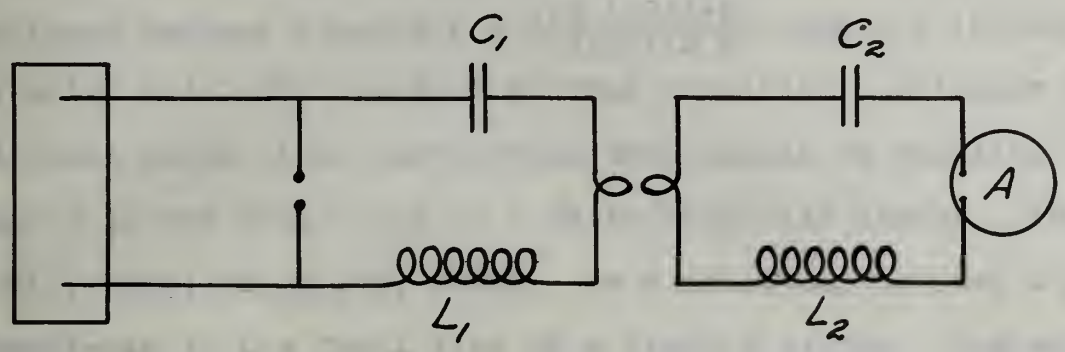


Figure 1

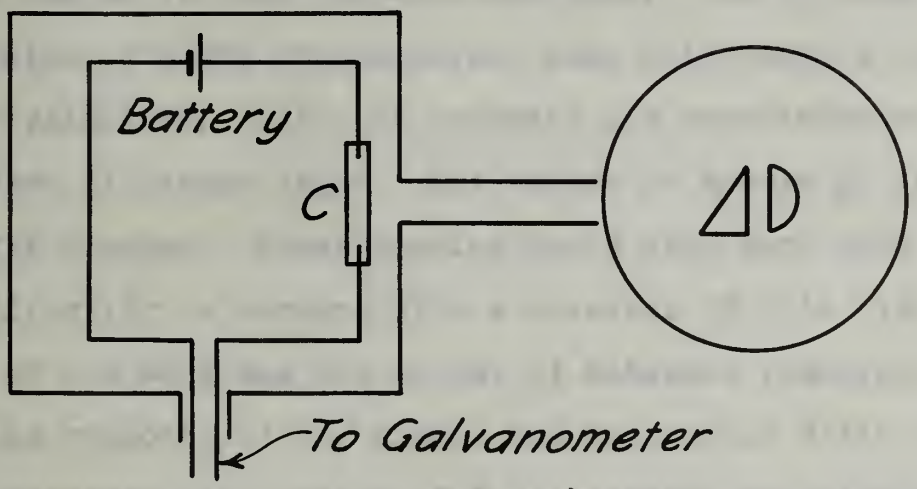


Figure 2

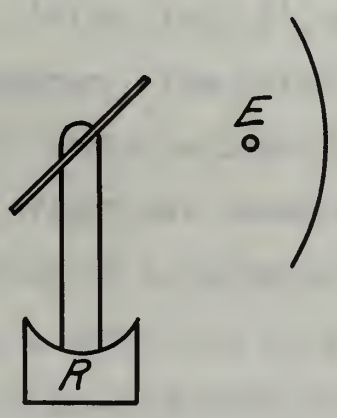


Figure 3

Figure 1

Figure 2

Figure 3

prism. By determining the angle of minimum deviation for plane waves of a given length the value of n may be calculated by means of the ordinary optical formula $n = \frac{\sin 1/2(A+D)}{\sin 1/2\ A}$, where A is the angle of the prism and D the angle of minimum deviation. Ellinger obtained plane waves of 60 centimeters wave-length by mounting a linear oscillator in the focal line of a Hertz parabolic mirror. (Wied. Ann. 46, p 511, 1892; 48, p 108, 1893). As a detector, he used a micrometer spark-gap in the focal line of a similar mirror. The angle of the prism was 3° 45'. When the axes of the mirrors made an angle of 30° with each other, sparks were produced at the micrometer gap. The refracting medium in this instance was water, and Ellinger concluded that the value of n was approximately that which should be expected from other relations. Later he repeated his experiment with alcohol, using a prism of larger angle. His values of k were 81 for water and 24.8 for alcohol. These results would seem very good considering the difficulty in working with a detector of this kind. One disadvantage of his work was the amount of material required for a test, as the capacity of the prisms was upwards of fifty liters.

Lampa obtained plane waves of 8 millimeters wave-length by using a small Righi oscillator in the focal line of a small parabolic cylinder of sheet metal. (Wied. Ann. 61, p 79, 1897). As a detecting device, he used a coherer. The prism and mirror were mounted on a circular table which could be rotated about a vertical axis (Fig.2) until a maximum effect was observed on the coherer, C, as indicated by the deflection of a galvanometer. The coherer, battery and leads were placed in a metal box to the side of which a metal tube was soldered. Waves passing down this tube and through the opening in the side of the box could act upon the coherer. Otherwise

the detecting system was protected by metal covering in order to eliminate stray effects. The aperture of the mirror was stopped down to a narrow vertical slit in order that only a narrow beam might be refracted. Inaccuracy in this work was due to the difficulty in obtaining settings because of the irregular action of the coherer, especially for smaller values of n.

2. Reflection Methods. Cole used a wave-length of 5 centimeters in determining the reflecting power of metallic surfaces and of dielectric surfaces. (Wied. Ann. 57, p 290, 1896). Using Fresnel's formulas for rays polarized parallel to the plane of incidence and for rays polarized perpendicular to the plane of incidence, two values of n were calculated which agreed very well. The mean of the two values was used in calculating k. Cole's apparatus consisted of a specially designed Righi oscillator, E, mounted in the focal line of a cylindrical mirror of sheet metal (Fig.3). As a detector a small linear oscillator was used in the focal line of the cylindrical mirror R. This linear oscillator consisted of two narrow strips of metal foil, each one centimeter long, and mounted end to end five millimeters apart. The near ends were connected to a nickel-iron thermal element of the Klemencic type which was included in a galvanometer circuit. (Ann. 45, p 62, 1892). By means of a wooden arm pivoted at the center of curvature of the transmitting mirror, the receiving mirror could be rotated into any desired position. Both mirror units could be removed from this arrangement and so mounted that reflections from the surface of a liquid in an open vessel could be measured. In this method, special care was necessary in order to protect the thermal element from all effects other than those due to the waves coming into the receiving mirror.

3. Interference Methods. The first interference method was that of von Lang. (Wied. Ann. 57, p 430, 1896). The arrangement of his apparatus is shown in Fig.4. The oscillator was of the Righi type. Two small spark balls placed in the sides of a small trough containing petroleum formed a gap about one millimeter long. Outside of the trough two larger balls were placed so as to add two air gaps, the whole constituting a linear oscillator which provided waves of about 44 millimeters wave-length. The interference apparatus consisted of a system OBCDE made up of metal tubing or of cardboard tubing covered with tinfoil. Two sets of tubes were used. The metal tubes were three centimeters in diameter, but later cardboard tubes six centimeters in diameter were used. The branch tubes C and D were long enough so that each could be drawn out twenty centimeters and thus increase the path from O to E by forty centimeters. Waves entering the slit at O divided at B, passed around through C and D to E and into the metal box containing the coherer and its galvanometer connections. With the tubes C and D of equal length a minimum effect was produced upon the coherer. Upon lengthening the path C, for instance, the action upon the coherer increased until it became a maximum for a path difference of one half wave-length. With the plane of the tube system vertical, the lower tube was filled with the substance to be tested. The upper tube was pulled out until two successive maxima had been observed. The increase in path between the two maximum positions gave the wave-length in the substance. The ratio of the wave-length in air to the wave-length in the substance gave the index of refraction n, from which k was calculated. The slit at O and the oscillator were placed vertically when the plane of the tubes was horizontal in order not to decrease

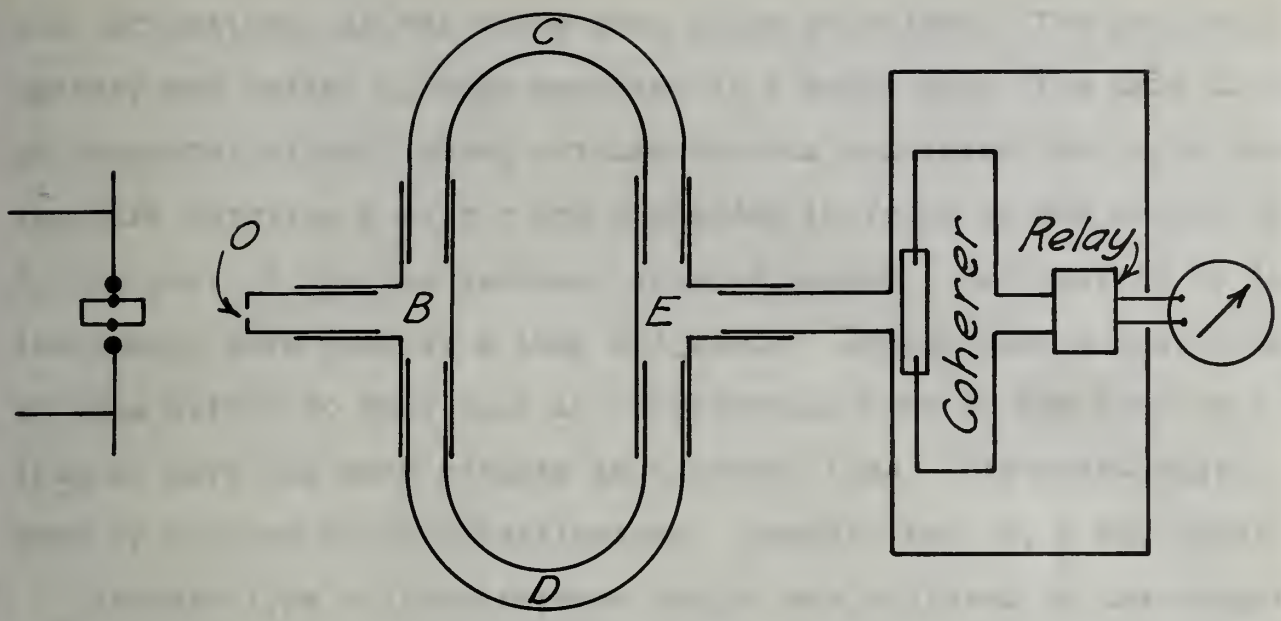


Figure 4

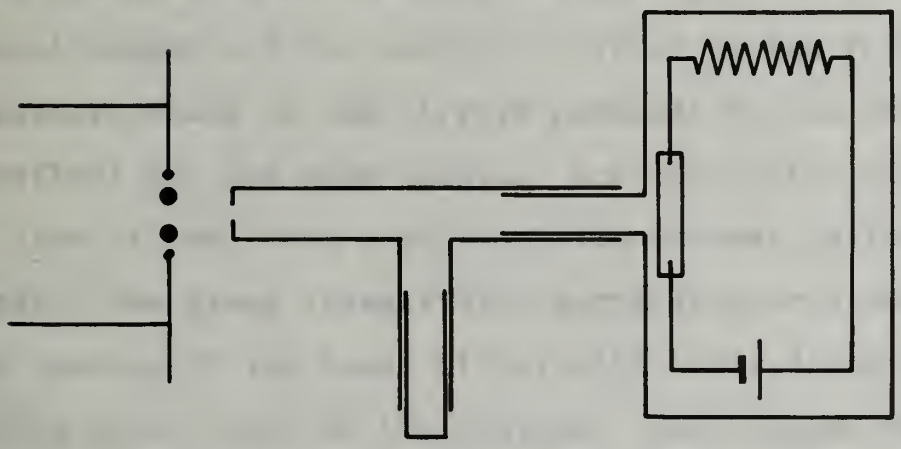


Figure 5

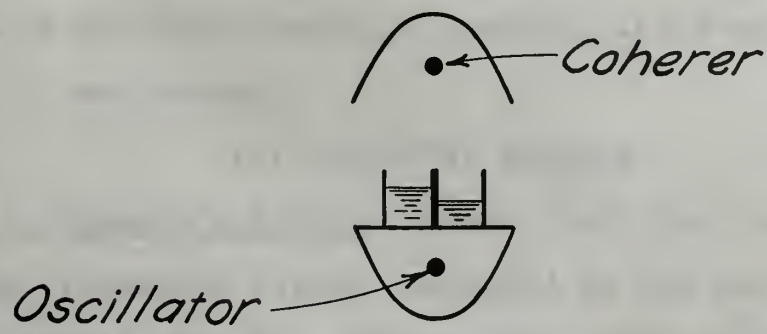


Figure 6

Figure 4

Figure 5

Figure 6

the reflections, as the waves were plane polarized. The coherer, battery and relay, R, were enclosed in a metal box, The part of the galvanometer circuit shown outside the box consisted only of a steel armature carrying a mirror and suspended in front of the magnet of R, the part of the box between being of copper. Deflections of this instrument were read by a lamp and scale. Becker used a modification of this method to show that an interference tube in the form of a T (Fig.5) gave the same results as a branch tube. The wave-length used by him was about 9 centimeters. (Drude's Ann. 8, p 22, 1902).

Another type of interference method was employed by Kossonogoff. (Phys.Zeit. 3, p 207, 1902). The arrangement of his apparatus is shown in Fig.6. Two parabolic cylinders were mounted one above the other. The aperture of each mirror was 10 x 10 square centimeters and the focal length 1.5 centimeters. Strips of tin foil were placed on the cardboard walls of the mirrors parallel to the focal lines in order to reflect but one wave-length. The oscillator was mounted in the focal line of the lower mirror and the coherer in the focal line of the upper. Two glass vessels with perpendicular sides were placed across the opening of the lower mirror with their dividing walls in line with the focal lines of the mirrors. The liquids tested were placed in the vessels and their surface levels adjusted until a minimum effect was observed at the coherer. The wave-lengths were calculated by the Boltzmann-Righi method, and ranged from 2 centimeters to 10 centimeters.

III CAPACITY METHODS

1. J.J. Thomson's Method. (Proc. Roy. Soc. Lond. 46, p 292, 1889). J.J. Thomson's method consisted in the use of a single high frequency circuit (Fig.7). Two parallel metal plates 30 centimeters

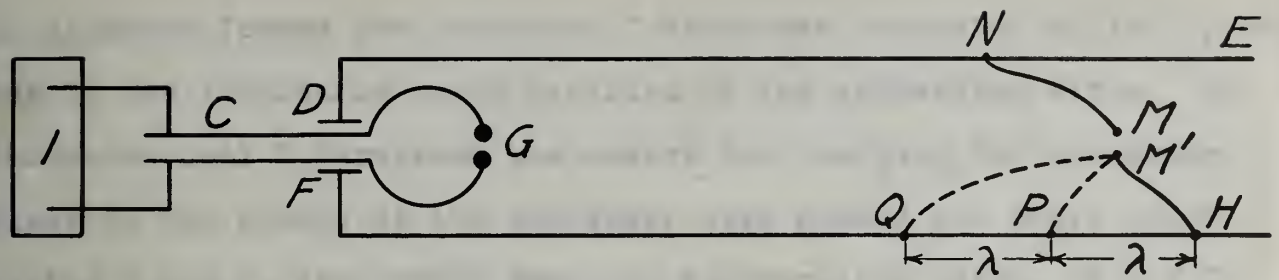


Figure 7

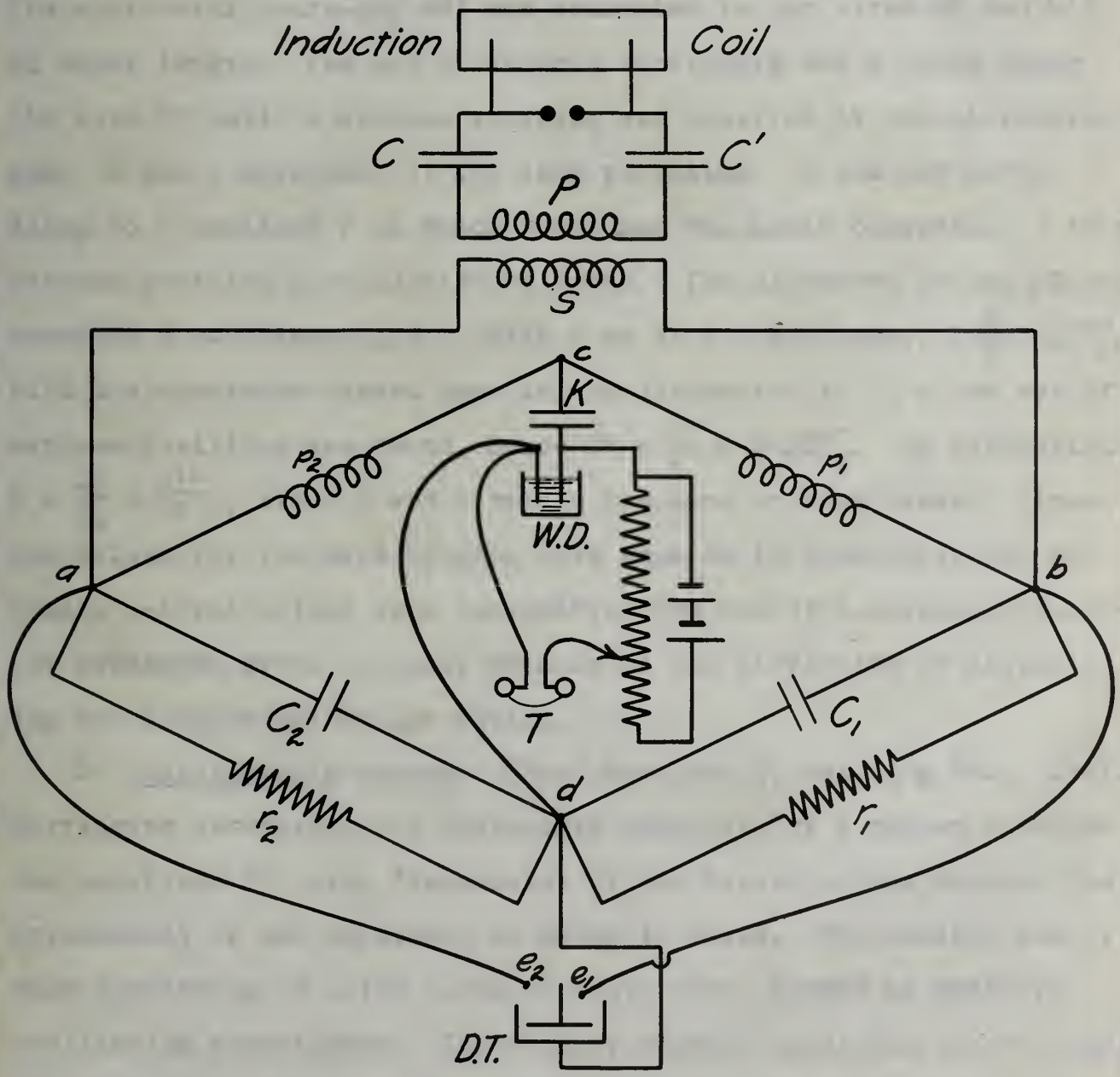


Figure 8

Figure 7

Figure 8

in diameter formed the condenser C which was connected to the spark-gap G, the inductance being provided by the connecting wires. An induction coil I furnished the energy for charging the condenser. Close to the plates of the condenser were placed two small metal plates D and F, from which were led two parallel wires, DE and FH, each about twenty meters long. By means of these wires the wave-lengths emitted by the oscillating circuit were measured directly. The micrometer spark-gap MM' was connected to two wires MN and M'H of equal length. The end H was held stationary and N moved along the wire ED until a minimum sparking was observed at the micrometer gap. H and N were then at the same potential. H was now moved along to a position P at which a minimum was again observed. A third minimum position Q could also be found. The distances HP and PQ represented a wave-length, λ. With C as an air condenser, $T=\frac{\lambda}{V}=2\pi\sqrt{LC_a}$. With the substance tested used as the dielectric in C, a new set of minimum positions was found, hence $T' = \frac{\lambda'}{V} = 2\pi\sqrt{LC_s}$. By definition $k = \frac{C_s}{C_a} = \frac{\lambda'^2}{\lambda^2}$, since L and v remain the same in both cases. Since the values for the wave-lengths were squared in calculating k, accurate determinations were necessary. The use of a spark-gap detector prevented great accuracy because of the difficulty of determining the minimum sparking position.

2. Harrington's Method. (Phys.Rev. Vol.8, Ser.2, p 581, 1916). Harrington determined the dielectric constants of a number of aqueous solutions for high frequencies by the Nernst bridge method. The arrangement of his apparatus is shown in Fig.8. Two coils, P and S, each consisting of a few turns of heavy wire, formed an ordinary oscillation transformer. The primary circuit consisted of two equal capacities C and C' in series with a spark-gap and the primary P of

the oscillation transformer. The terminals of the secondary coil S were connected to the terminals a and b of the bridge network. p_1 and p_2 were inductances each 9 centimeters in diameter and 10 centimeters long and containing 50 turns. C_1 and C_2 were variable plate condensers, r_1 and r_2 non-inductive resistances, and DT the test condenser. By connecting the free terminal of DT to e_1 or e_2, the test condenser could be put in parallel with either C_1 or C_2 as desired. Between the points c and d of the network, it was necessary to use a detector instead of the usual galvanometer or telephone. This consisted of an electrolytic detector WD in series with a large condenser of 3.2 microfarads. Shunted around the detector was a telephone receiver T with a suitable potentiometer arrangement. When the bridge was properly balanced there was no sound in the telephone. In determining the dielectric constant of a liquid four capacity determinations were made. The capacity of DT was determined for two positions of the adjustable upper electrode. If $s_1 - s_2$ represented the change of capacity with air, $S_1 - S_2$ the change with the liquid as dielectric, then $k = \frac{S_1-S_2}{s_1-s_2}$. By taking these differences, the effect of surrounding bodies upon the capacity of the test condenser was eliminated.

3. Resonance Methods. (A) Maximum Methods.

a. Thwing's Method. (Zeit. fur Phys. Chem. 14, p 286, 1894). Thwing was the first to employ a resonance method. The method was suggested by Professor Hertz, and was used by Thwing in determining the dielectric constants of a large number of substances. Fig.9 shows the arrangement of his apparatus. The primary circuit consisted of a variable condenser C_p in series with a spark gap G, the inductance being furnished by the connecting wires, which were in

the form of a square 60 centimeters on a side. Separated from the primary circuit by a distance of 15 centimeters, the secondary circuit consisted of a variable plate condenser C_s, for which the test condenser could be substituted, connected in series with a hot-wire galvanometer, S. The connecting wires were in the form of a square of the same dimensions as in the primary, so that the inductance was the same in both circuits.

With the test condenser in the secondary circuit the primary circuit was "tuned" to resonance with the secondary by adjusting C_p until a maximum deflection of the hot-wire galvanometer was obtained. Then the calibrated variable condenser was substituted for the test condenser and adjusted until the deflection was again a maximum. Evidently the capacity of the condenser C_s equals C_a for the test condenser. The test condenser filled with liquid was again placed in the secondary and the primary again brought to resonance with the secondary. The test condenser was again replaced by the variable condenser C_s, which was adjusted until the secondary was in resonance with the primary. Calling this value of the variable condenser C_1,

$$k = \frac{C_1}{C_a} .$$

The hot-wire galvanometer was constructed as indicated in Fig.10. Two German silver wires each .12 mm. in diameter and 30 centimeters long composed the heating element. One end of each wire was soldered to the side of a small steel bar cd ,86 millimeter in diameter, upon which was mounted a small mirror m. A steel wire .25 millimeter in diameter connected the lower end of the bar to a rigid support e, and a similar wire connected the upper end of the bar to a torsion head T. The rotating system was mounted vertically and the whole system made up one side of the secondary circuit. By twisting the

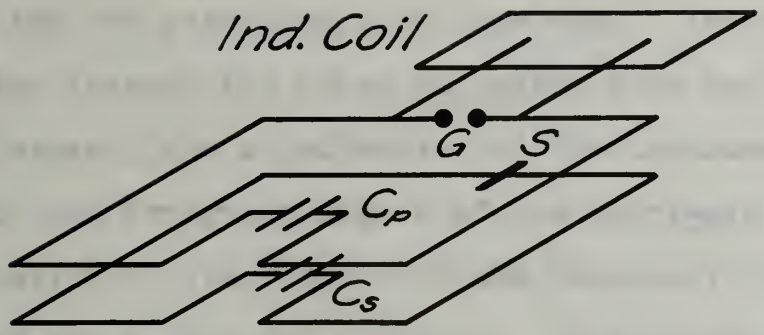


Figure 9

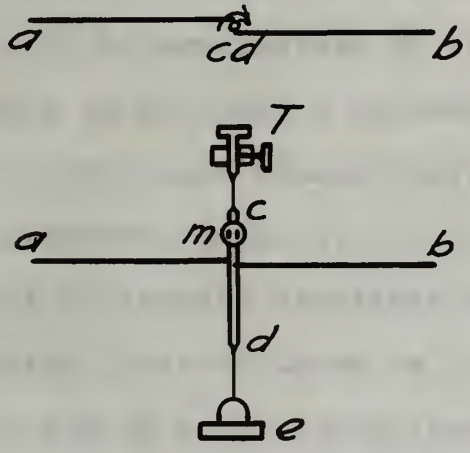


Figure 10

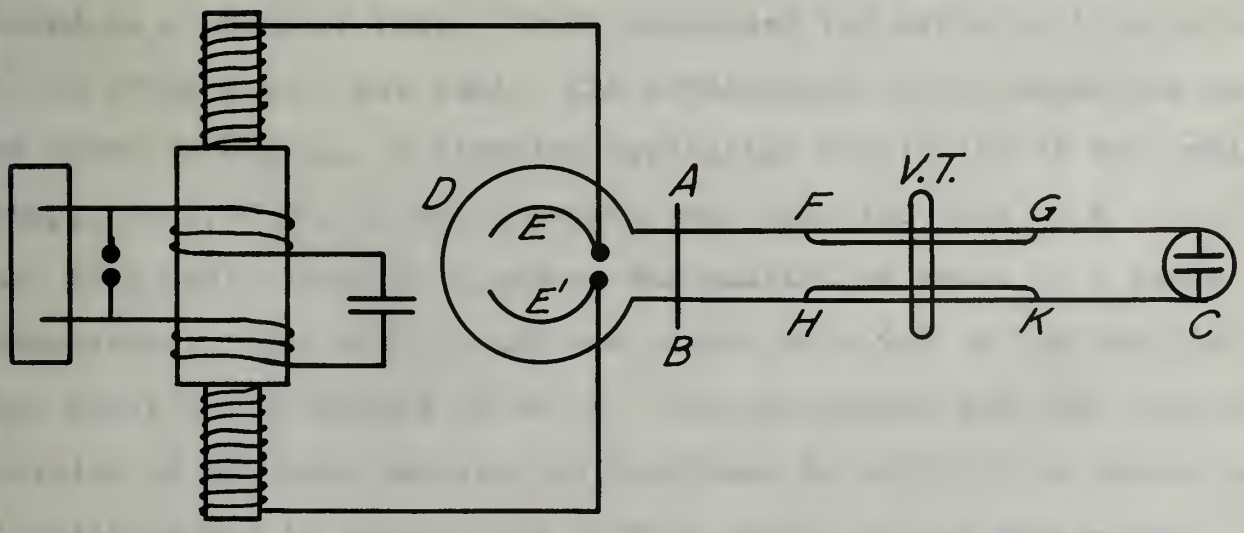


Figure 11

torsion head, the two wires could be made taut. The oscillating currents passing thrcugh the wires ab heated them sufficiently to cause them to expand, and a deflection of the instrument resulted. In this method, the irregular action of the spark-gap caused sufficient fluctuation of the current in the secondary circuit that a "fluttering" of the mirror was produced. This necessitated the use of a lamp and scale instead of a telescope for reading the deflections. Also the length of the German silver wires used made it necessary to protect the apparatus from air currents as far as possible. The resistance of 60 centimeters of German silver wire of the size used is upwards of 12 ohms, a circumstance which would decrease the sharpness of resonance considerably.

b. Drude's Second Method. (Ann. 8, p 336, 1902). Drude determined a large number of dielectric constants by a number of similar methods, finally adopting a method known as Drude's Second Method, which has been widely used by other investigators. This method made use of resonance circuits in a parallel wire system, resonance being obtained by varying the inductance until a maximum glow was produced in a Geissler tube. Drude concluded the error in this method to be about 2 or 3 per cent. The arrangement of his apparatus was as shown in Fig.11. A Blondlot oscillator consisting of two small copper rods, E E', of equal length bent into the form of a circular arc five centimeters in diameter was excited by means of a Tesla transformer. The circuit ABD was placed parallel to the oscillator and about 3 millimeters below it. The oscillator and the circular portion of ABD were immersed in petroleum in order to increase the insulation and to insure more uniform action of the spark-gap. AB was a wire bridge which was placed in a fixed position just outside

the oil container. Two parallel wires extended to the right, their extremities being connected to the small condenser C. The portions FG and HK of the parallel wire system consisted of telescoping tubes in order that the circuit ABC might be lengthened or shortened easily and quickly. Oscillations were induced in ABD by the oscillator EE'. The bridge AB constituted the coupling between ABD and ABC. When resonance was secured between ABD and ABC, the distance from AB to the extremities of the wires was one fourth of a wave-length, since the bridge must lie at a node of potential and the condenser at a loop. The system was calibrated by using liquids of known dielectric constant in C in order that resonance settings might be made without making great changes in the length of the circuit ABC. The ratio of the squares of the wave-lengths gave the ratio of the two capacities, from which values k was calculated. Drude emphasized the fact that the oscillator should be close to ABD because oscillations of two periods were set up in ABD, and for close coupling the shorter wave-length oscillations would be much weaker than the other. With the distance given, the ratio of the periods was approximately 2 to 1 so that when AB was at a node of the longer wave, it was at a loop of the shorter. In this way he avoided errors that might be made at times by tuning the circuit ABC to different periods.

c) Niven's Method. (Proc. Roy. Soc. Lond., Ser.A 85, p 139, 1911). Niven modified Thwing's method by placing the test condenser in the primary circuit and by using a calibrated secondary circuit. (Fig.12). The primary circuit consisted of the test condenser C in series with a rectangular inductance L and a spark-gap G, energy being supplied by an induction coil. The secondary circuit was a

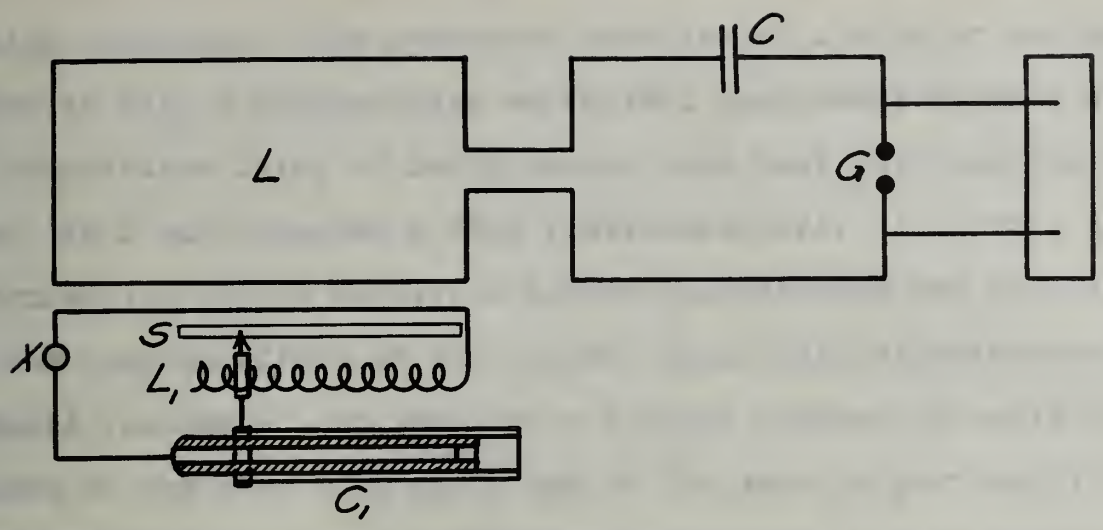


Figure 12

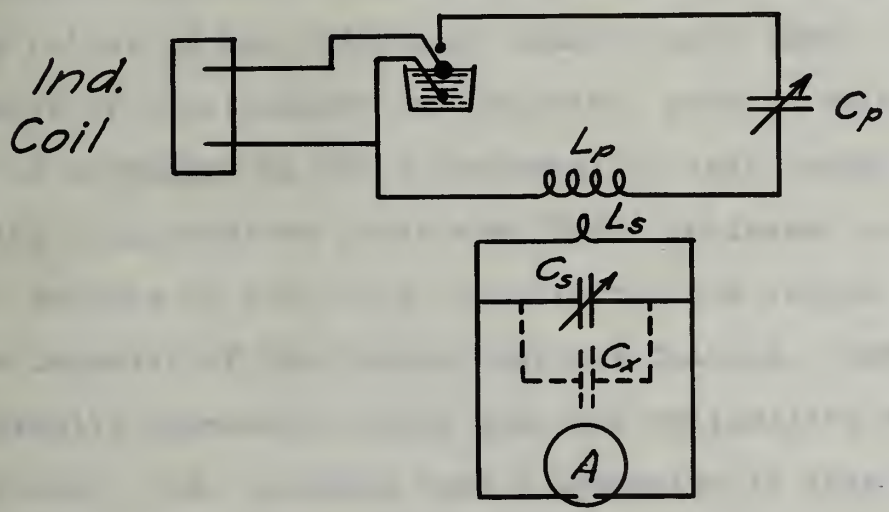


Figure 13

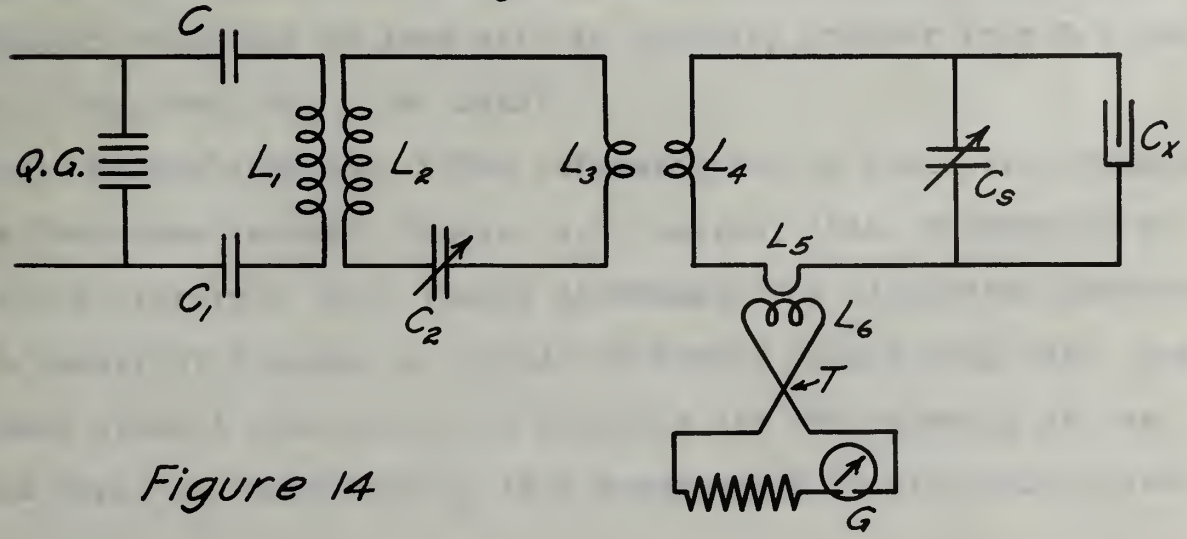


Figure 14

Fleming cymometer. The cymometer consists of a tubular condenser C_1 in series with a single-layer solenoidal inductance of bare wire, the connections being of heavy copper rods bent into rectangular form. At X was inserted a fine resistance wire. A thermal element in connection with a sensitive mirror galvanometer was used to measure the heating effect of the current upon this resistance and thus indicate resonance. By means of a sliding contact L_1 could be increased at the same time as C_1 and in the same proportion. The scale S was calibrated to read directly wave-lengths in feet and in meters, frequency, and values of the oscillation constant $\sqrt{LC}$. Niven used the oscillation constant values in determining the capacities of C, computing values of the inductance used in each case. In working with liquids of considerable conductivity, such as water and alcohol, he found it necessary to put a condenser of very large capacity in series with C in order to force the liquid condenser into oscillations. Because of the large capacity of this series condenser, the effective capacity of the circuit was not changed. The accuracy of Niven's results depended largely upon the reliability of the cymometer settings. S.H. Anderson used a cymometer in this laboratory in some high frequency investigations, and his conclusion was that the cymometer could not be read with an accuracy greater than 2.1 per cent. (Phys.Rev. 34, p 34, 1912).

d) Warner's Method. ("The Determination of Dielectric Constants by a Resonance Method", Thesis, A.M. degree, 1914, University of Illinois Library). E.H. Warner determined the dielectric constants of a number of liquids by a modified Thwing method (Fig.13). The primary circuit consisted of a variable air condenser C_p of the Korda type, an inductance L_p 14.5 centimeters in diameter of six

turns of wire, and a spark-gap, all in series. The gap was specially designed to provide uniformity of action. Using one large spark ball and two smaller ones above and below, two gaps in series were formed, the lower gap being immersed in oil. The action of this gap proved quite uniform. The secondary circuit consisted of an inductance of one turn, L_s, in series with a calibrated Korda condenser C_s. A Duddell thermal galvanometer was connected in parallel with C_s. The two circuits were adjusted to resonance with the condenser C_s set at some value near its maximum. The test condenser C_x was then connected in parallel with C_s and resonance was again obtained by reducing the value of C_s. The amount C_s was reduced was evidently the capacity of C_x. The ratio of the values obtained for C_x with and without the liquid gave the dielectric constant directly. Warner concluded that his results were in error from one to two per cent. The frequency used was computed to be approximately seventy millions.

e) Banneitz' Method. (Annalen 59, p 239, 1919). The method used by Banneitz is indicated in Fig.14. In this arrangement, an intermediate circuit was used, making three circuits to be adjusted to resonance. The primary circuit consisted of a thirty-plate quenched spark-gap, two Leyden jars C and C_1 of 600 centimeters and 740 centimeters capacity respectively, and an inductance L_1. The intermediate circuit consisted of the inductances L_2 and L_3 and a variable capacity C_2. The third circuit, used as the measuring circuit, consisted of the inductances L_4 and L_5 in series with a calibrated variable condenser C_s. The test condenser C_x was placed in parallel with C_s and its capacity determined by substitution, as in Warner's method. To detect resonance an auxiliary circuit was

coupled to the measuring circuit by means of the inductances L_5 and L_6. The terminals of L_6 were connected to a thermal element T. The thermal effects at T were indicated by a sensitive galvanometer. The galvanometer circuit proper contained a resistance in series, also two choke coils not shown in the sketch. Energy was furnished to the primary circuit by means of a resonance transformer supplied by a 500-cycle generator driven at a constant speed. The use of a quenched spark permitted close coupling between the first two circuits and feebly damped oscillations were obtained in the second circuit. The coupling between the intermediate circuit and the measuring circuit was made very loose in order to reduce the reaction between the two to a minimum, and to increase the sharpness of resonance. The test condenser consisted of three parallel plates, the outer two of which were connected together and grounded. Also, the condenser C_s was connected so that its fixed plates were grounded by the same connection. This ground connection served to protect the two condensers from the capacity effects of surrounding bodies. High accuracy was claimed for this method. Capacities within a range of 0 to 800 centimeters could be measured to 0.2 centimeters. As a test, Banneitz determined the dielectric constant of petroleum, which he claimed to be accurate to 0.04 per cent. The wave-length as measured by a wave-meter was 158 meters.

(B) Null Methods.

a) Ferry's Method. (Phil.Mag. (5), 44, p 404, 1897). The method of Ferry (Fig.15) was the first null method to be used in high frequency determinations of the dielectric constant. He used three loops of wire in the form of squares, all of the same dimensions and spaced approximately ten centimeters apart. In one side

of each square he placed a condenser. A spark-gap was placed in the opposite side of the inner square, and that circuit was used as the primary. Since all three inductances were equal, these circuits were in resonance when the capacities C_1, C_2, and C_3 were all equal. The bolometer principle was employed to detect resonance. The sides a_1b and a_2d of the outer squares were made from equal lengths of No.36 iron wire whose resistances were made equal to one part in one hundred thousand as checked by the Carey Foster bridge. The resistances b' and d' were made equal and wound side by side on a brass rod. With a battery connected between the points a and c, and a galvanometer between b and d, a Wheatstone network (Fig.16) was formed, and no deflection would result if the resistances aa_1b and aa_2d were equal at one temperature and had the same temperature coefficient of resistance, provided the arms bc and dc were also equal for all temperatures. To adjust the apparatus, the condenser C_1 was disconnected so that no currents would flow through a_1b except from the battery B. By varying C_2 the other two circuits were adjusted to resonance as indicated by a maximum deflection of the galvanometer. Then C_3 was taken out of its place and put into the other secondary, which was adjusted until the same deflection was obtained, in the opposite direction however. This adjustment was made by varying the distance between the two oscillating circuits until the deflection reached the desired value. C_3, which was the test condenser, was left in this circuit, and C_1, which was a standard variable condenser, was put into the other secondary. Varying C_1 until the galvanometer deflection was reduced to zero, the three circuits were in resonance, and the capacities of C_1 and C_3 were equal in value. While the value of k was determined finally from the ratio of two

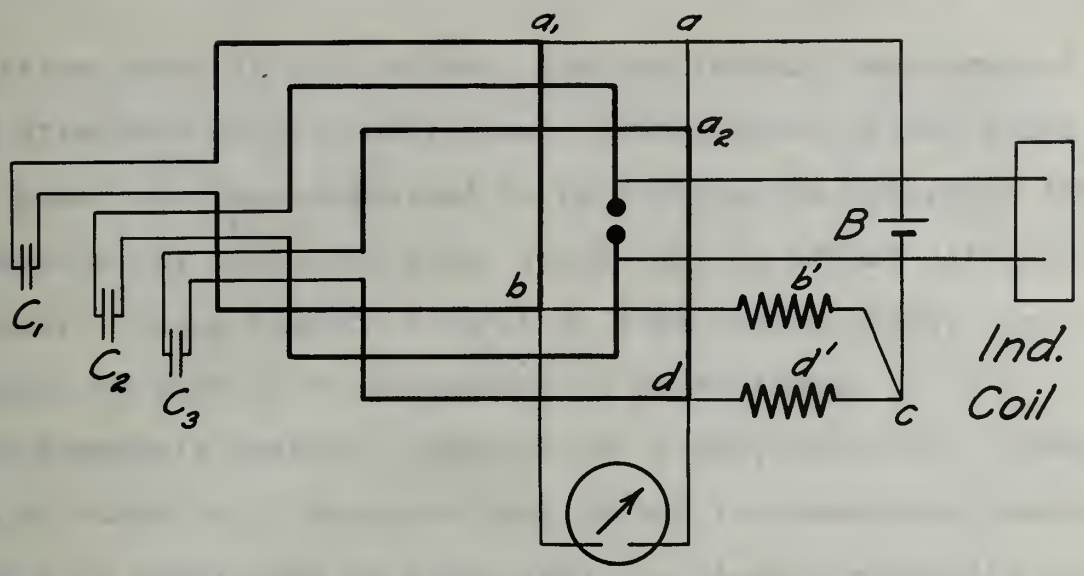


Figure 15

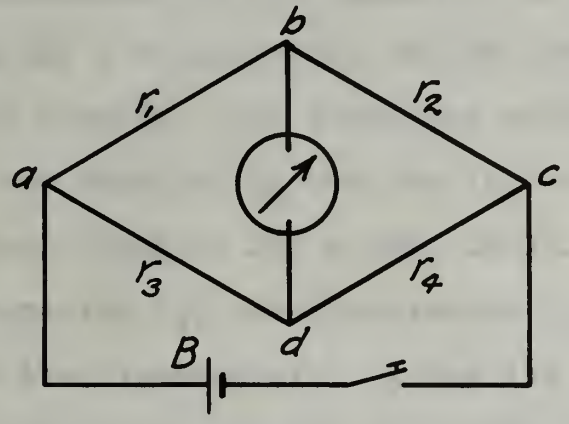


Figure 16

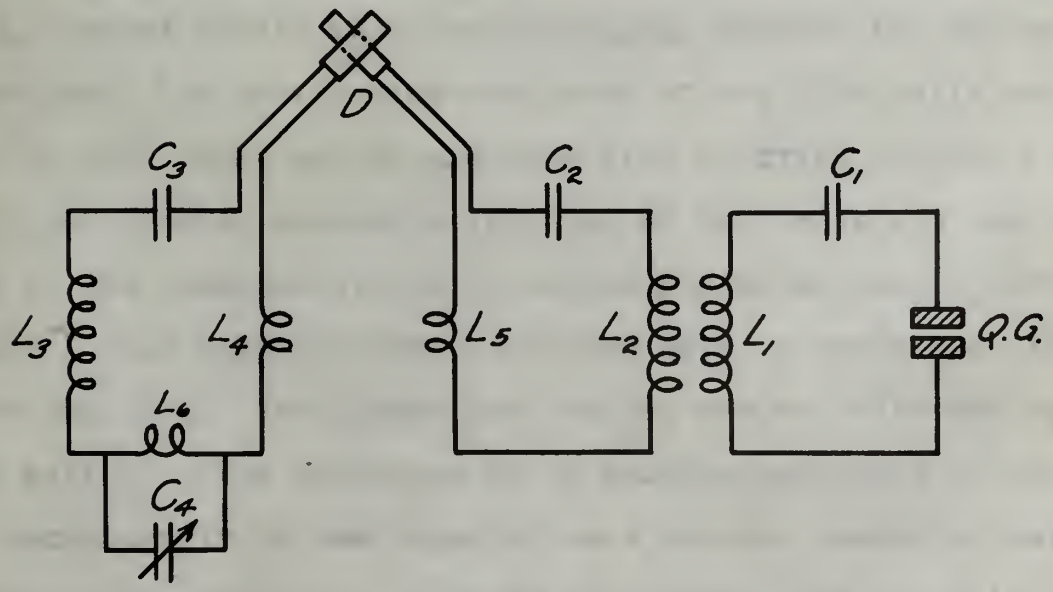
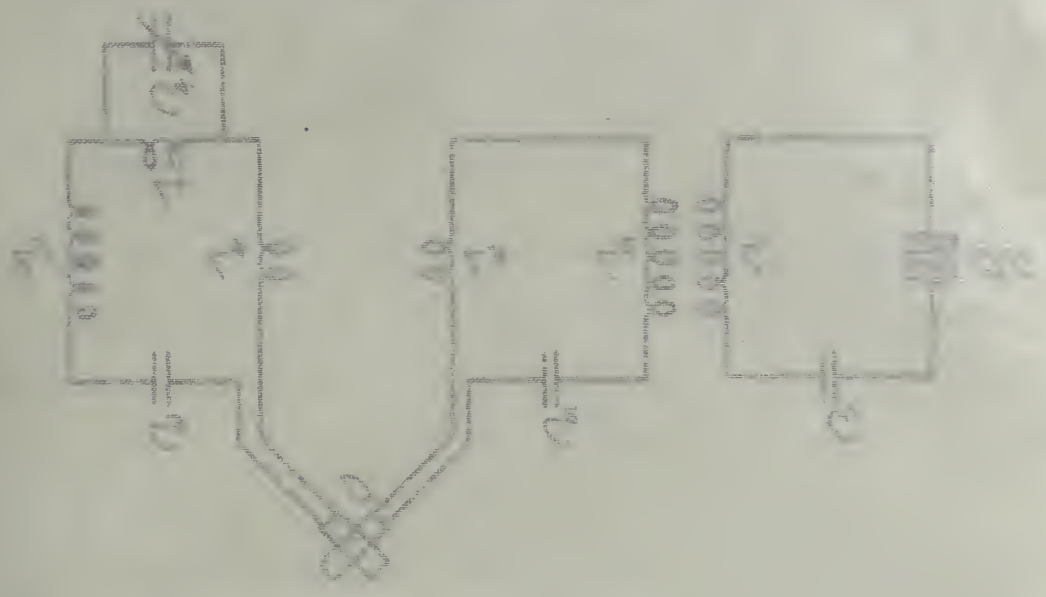
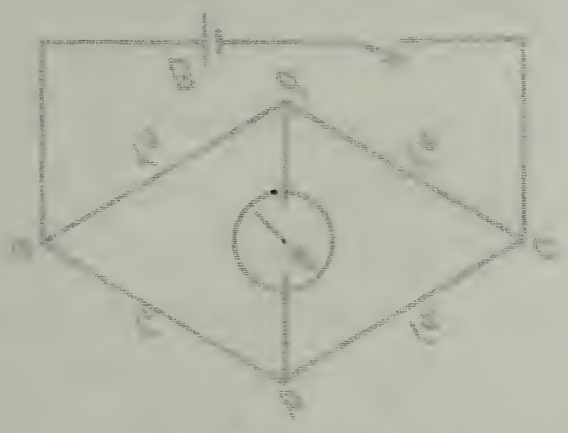
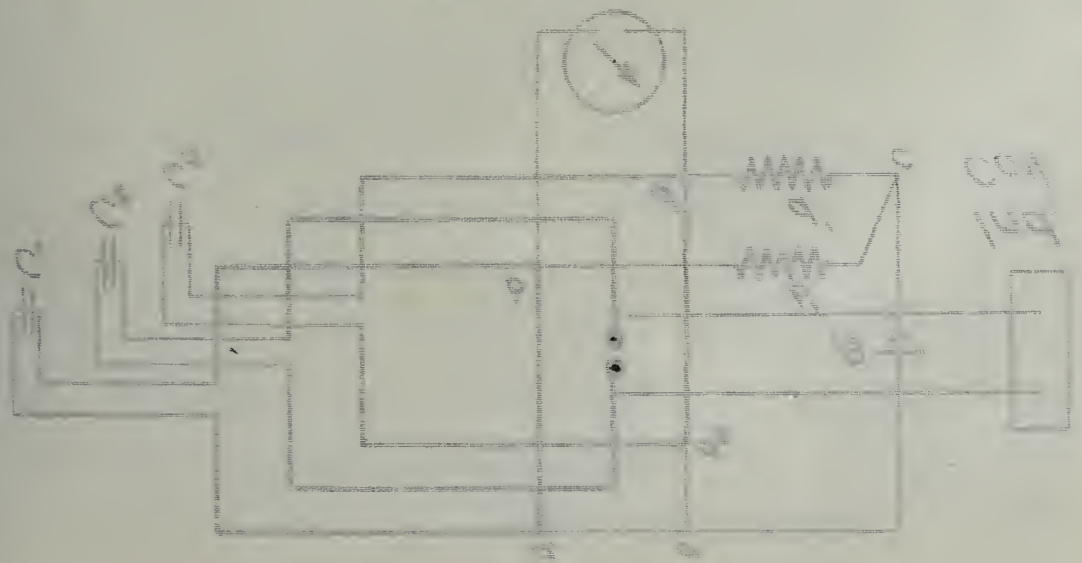


Figure 17







capacities found in this manner, the preliminary measurements and their attendant errors contributed to the errors of the final result. Also, great care was exercised in protecting the apparatus from air currents or any influence which would tend to affect different parts unequally. Using Thomson's parallel wire method, Ferry computed the frequency he used to be approximately 33 millions.

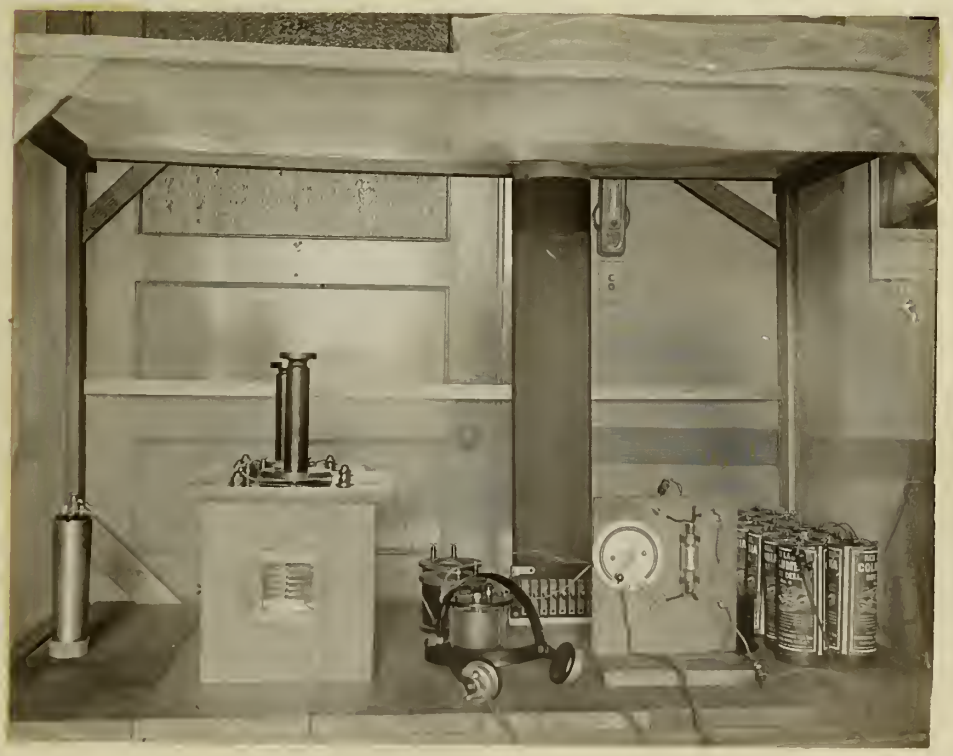
b) Rohmann's Method. (Annalen 34, p 979, 1910-11). Rohmann devised an exceedingly accurate null method for measuring changes in capacity in order that he might test the Clausius-Mossotti relation for gases. The arrangement of his apparatus is shown in Fig.17. A quenched spark-gap QG, a capacity C_1, and an inductance L_1 in series made up the primary circuit. One secondary consisted of the inductances L_2 and L_5, the capacity C_2 and one coil of a Mandelstam-Papalexi dynamometer. (Annalen 33, p 490, 1910). The other secondary consisted of the capacity C_3, the inductances L_3, L_4, and L_6, and the second coil of the dynamometer. C_3 was the test condenser. The inductance L_6 in connection with the capacity C_4 formed an auxiliary circuit. C_4 was a calibrated variable condenser. The inductances L_4 and L_5 served merely as a loose coupling between the two secondary circuits. The dynamometer consisted of two flat coils perpendicular to each other and an aluminum ring carrying a small mirror. The ring was mounted coaxially with one of the coils and was deflected by the dynamometer action of the currents passing through the coils. When both secondary circuits were in resonance, the deflection was zero. The dynamometer action was not affected by irregular action of the spark-gap or by varying amplitude of the currents, consequently it was superior to a current measuring device for determining resonance. Rohmann calculated that capacity changes

in the condenser C_3 could be measured with an accuracy of one part in one hundred thousand. For a change of the measuring condenser setting from C_4 to C_4', he showed that the change in C_3 denoted by δC_3 was expressed by the relation

$$\frac{\delta C_3}{C_4 - C_4'} = -\left(\frac{L_6}{L_3}\right)^2 .$$

This method is admirably adapted to the determination of the dielectric constants of gases. To apply it to liquids or solids, however, an enormous capacity would be needed in the condenser C_4. The frequency of this system as calculated by Rohmann was approximately two millions.

Apparatus used in Primary Circuit



Apparatus used in Test Circuit

E X P E R I M E N T A L

IV INTRODUCTION

The purpose of this investigation was to develop a method of determining the dielectric constants of liquids for high frequencies. Such a method should be accurate, but simple and as nearly direct-reading as is practicable. The arrangement should be such that only a small quantity of liquid would be needed in making a determination and that the necessary adjustments might be made easily and quickly. Also, it should be possible to study the liquid under varying conditions of temperature and pressure.

A resonance method seemed best adapted to meet these conditions. The accuracy of resonance methods heretofore developed has been limited by the variations of frequency and amplitude of the currents in the primary circuit due to irregular action of the spark-gap. Another limitation has been the inability to detect resonance with high accuracy. In developing this method it was decided to make use of a three-electrode vacuum tube, or audion, as a source of high frequency currents. Such a tube when properly connected in an oscillatory circuit generates oscillations of constant amplitude and of a frequency determined by the capacity and inductance of the circuit. Instead of the usual methods of detecting resonance it was decided to employ a means of direct comparison of frequencies. This was accomplished by making use of the heterodyne principle, or principle of beat reception, commonly used in radio work for the reception of signals transmitted by undamped waves.

V METHOD

The method consisted in the use of two loosely coupled oscillatory circuits, one of which was operated at a constant frequency

during a test and therefcre may be considered as the primary circuit. The second circuit contained a calibrated variable condenser used as a standard, and was sc arranged that the test condenser could be connected in parallel with the standard, and its capacity determined by substitution.

The connections of the primary circuit are indicated in Fig.18.

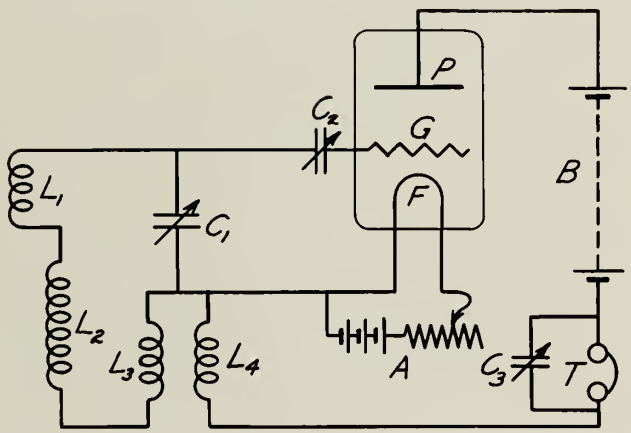


Fig.18

The inductances L_1, L_2 and L_3 in series with a condenser C_1 formed a closed oscillatory circuit. One terminal of the condenser C_1 was connected to the negative side of the filament F of the tube, and the cther to the grid G through a condenser C_2. The positive terminal of a battery B of about 24 volts was connected to the plate P. The negative terminal of the battery was connected to the negative side of the filament through high resistance telephone receivers T and an inductance L_4. A condenser C_3 was connected in parallel with the telephone receivers in order to provide a path of low impedance for high frequency currents. The filament was heated by current

from a six-volt battery A. E.H. Armstrong has shown that, with the coils L_3 and L_4 in the proper inductive relation and closely coupled, undamped oscillations of a frequency determined by the inductance and capacity of the closed circuit are maintained in the system. (Sci.Abs.18,Ser.B, p 494, 1915; abstract of article in Proc. Inst. Radio Engrs. 3, p 215, 1915).

The test circuit, Fig.19, was of the type originated by Dr.Lee

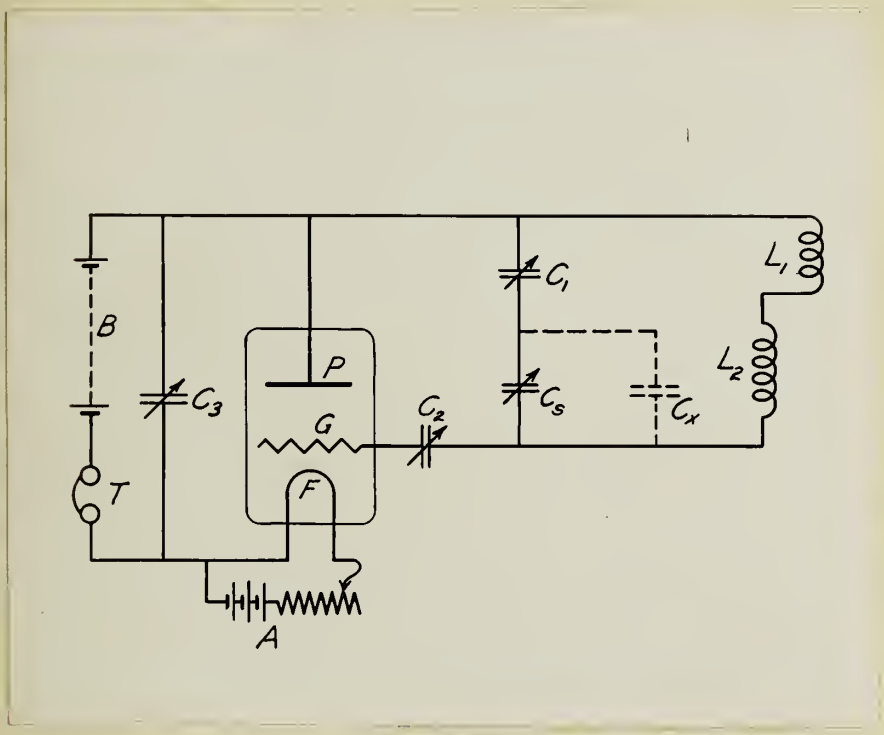


Fig. 19

DeForest and named by him the "ultraudion" circuit. (Elec.World,65, p 465, 1914). This arrangement differs from the first in that the closed circuit is connected between the grid and the plate, the plate itself providing the coupling between the grid and plate circuits necessary to maintain oscillations. The standard variable condenser C_s was connected in series with the condenser C_1. The coupling between the two circuits was effected by placing the coil L_1 of the test circuit in inductive relation with the coil L_1 of the primary circuit. In order that its frequency might not be affected

by the capacity of the body of the observer, the test circuit was enclosed in a grounded cage of wire netting. The condensers C_1 and C_s were provided with long handles of hard rubber so that settings could be made from outside the cage. The primary circuit was not enclosed as it was on the opposite side of the cage from the observer and sufficiently distant that it was not affected noticeably. The coupling coil L_1 of the primary circuit was placed in the cage, however, the leading-in wires being carefully insulated from the wires of the cage by rubber tubes.

For the capacity measurements necessary in determining the dielectric constant, the test condenser C_x was connected in parallel with the standard condenser. The effective capacity of the circuit was increased and the frequency of the oscillations therefore decreased. By reducing the capacity of the standard condenser by an amount equal to C_x, the frequency was restored to its former value. The calibration data of the standard condenser showed that the increase in capacity from the minimum to the maximum value was directly proportional to the scale reading. The capacity of C_x with air as the dielectric may be expressed by the relation $C_a = A(R_1-R_o)$, where R_o represented the initial scale reading, R_1 the reading after C_x was connected in parallel, and A the capacity per scale division. Similarly, the capacity of C_x with a liquid dielectric may be expressed by the relation, $C_b = A(R_2-R_o)$, where R_2 was the scale reading with the condenser C_x filled with liquid. Then by definition, the dielectric constant $k = \frac{C_b}{C_a} = \frac{R_2-R_o}{R_1-R_o}$.

The heterodyne principle was first used by Fessenden in receiving signals sent out by an undamped wave transmitter. (Elec.Rev. 60,p368, 1907). He constructed a special telephone receiver in which the

permanent magnet was replaced by a small electromagnet, the core of which was made up of a large number of iron wires one-thousandth of an inch in diameter. Mounted on the diaphragm, which was of mica, was a coil of fine wire. Oscillations induced in the receiving circuit were led through the coil on the diaphragm. Currents of a slightly different frequency furnished by a high frequency alternator were sent through the coil of the electromagnet. The action of the electromagnet upon the diaphragm coil caused the diaphragm to vibrate with a frequency equal to the difference between the frequencies of the currents. With the alternator frequency properly chosen the vibrations of the diaphragm produced an audible note in the telephone. The development of the oscillatory tube circuits for receiving made it possible to produce beats by the interference between the oscillations set up by the tube and the oscillations induced by the incoming waves. Beat reception depends upon a certain degree of detuning, since no beats are produced when circuits are in resonance. The application of the heterodyne principle to this method consisted in adjusting the inductance or capacity of the two circuits until a given beat frequency was noted in the telephones. The final adjustments for all readings were made by using the condenser C_s. The beat frequency was determined by tuning the note in the telephone to unison with that of a tuning fork having a frequency of 512. This was done by adjusting C_s until the acoustic beats produced between the telephone note and the tuning fork note disappeared. Since a setting could be made on either side of the resonance position, it was necessary to exercise care that all settings should be made on the same side of the resonance position.

VI MANIPULATION

In making a determination of the dielectric constant of a liquid the standard condenser C_s was set to some value near its maximum and the series condenser C_1 to some value near its minimum. The outer shell of the test condenser (Fig.20) was connected to one terminal

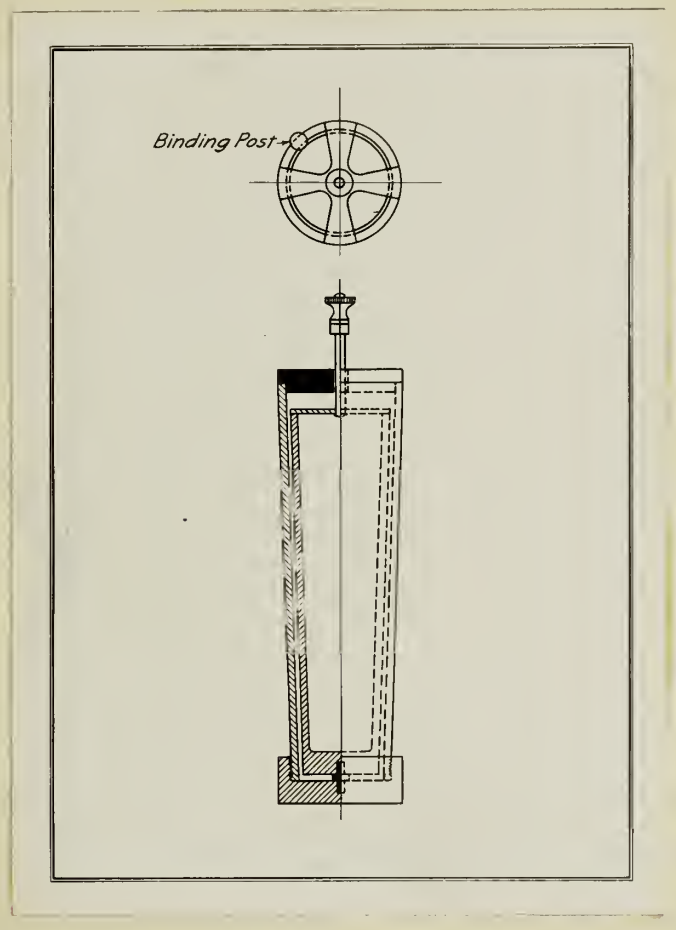


Fig. 20

of the standard condenser in order that its capacity due to surrounding objects might be a part of the effective capacity of the circuit for all readings. The circuits were then put in operation by adjusting the filament currents to their normal value, after which the primary circuit was adjusted until it was approximately in resonance with the test circuit. Returning to the test circuit, adjustments were made of C_1 and lastly of C_s until the beat note was in unison with that of the tuning fork. This setting of C_s was designated as

R_o. After connecting the inner cone so as to put the test condenser in parallel with C_s the capacity of C_s was reduced until the beat note was in unison with that of the fork. This reading of C_s was designated as R_1. The test condenser was then filled with the liquid and the capacity of C_s again reduced until the beat note was in unison with that of the fork. This reading of C_s was designated as R_2. The ratio $\frac{R_2-R_o}{R_1-R_o}$ gave the value of the dielectric constant directly.

Attempts to make determinations for distilled water, glycerine, acetone and absolute alcohol were unsuccessful. Since the standard condenser was not sufficiently large to permit determinations of dielectric constants larger than 4.35, a small condenser of known capacity was put in series with the test condenser for these trials. In each case the conductivity was such that the resultant capacity of the combination reduced to that of the series condenser and the tests failed. A one-microfarad condenser was added to the series combination with no change in the results. By keeping the condenser C_1 set for a small value, the change in the effective capacity produced by the addition of the test condenser to the circuit was only a few per cent, and the range of settings of the standard condenser over which a beat note could still be heard was rather large. In this way it was made certain that the circuit was tuned to the same frequency each time, and not to some overtone. This frequency was calculated to be approximately 75,000.

VII APPARATUS

The test condenser (Fig.20) was a conical condenser of the type used by Fleming and Dewar. (Proc. Roy. Soc. Lond. 61, p 299, 1897). The inner cone was 15.5 centimeters long, 3.23 centimeters in diameter at the bottom and 3.8 centimeters in diameter at the top. The

outer shell was 16.5 centimeters long, 3.43 centimeters in diameter at the bottom and 4.06 centimeters in diameter at the top. The inner cone was insulated from the outer shell and held concentric with it by a small hard rubber button at the bottom and a four-pronged hard rubber spider at the top. The base of the outer shell was removable so that the condenser could be taken apart and cleaned very easily. The parts had been turned from brass castings and were platinized to prevent corrosion by any of the substances used.

The standard variable condenser was an air condenser of the Korda type (Fig.21). It was one of two which had been built by Mr. Hays, the department mechanician, especially for measurement work. The plates were turned from brass castings to insure rigidity and special pains were taken in assembling them to make them as nearly parallel as possible and uniformly spaced. The plate system of each condenser was then mounted firmly between two slabs of white Colorado marble which had been boiled in paraffin to increase the insulation. The scale was cut to read from zero to 180 and was provided with a vernier attachment, permitting accurate readings to one-tenth degree. The box provided for each condenser was lined with sheet metal. By means of a binding post on the outside of the box, this metal lining could be grounded when the condenser was being used and outside influences thus eliminated. The calibration curve for the condenser used as the standard is shown in Fig.22. The data for this curve was taken by Mr. S.F. Tanabe, in February 1917, and the end points checked one month later. The capacity was determined by the divided charge method, and readings were taken for settings at intervals of ten degrees from zero to 180 degrees. The condenser in series with the standard was of the same construction as the

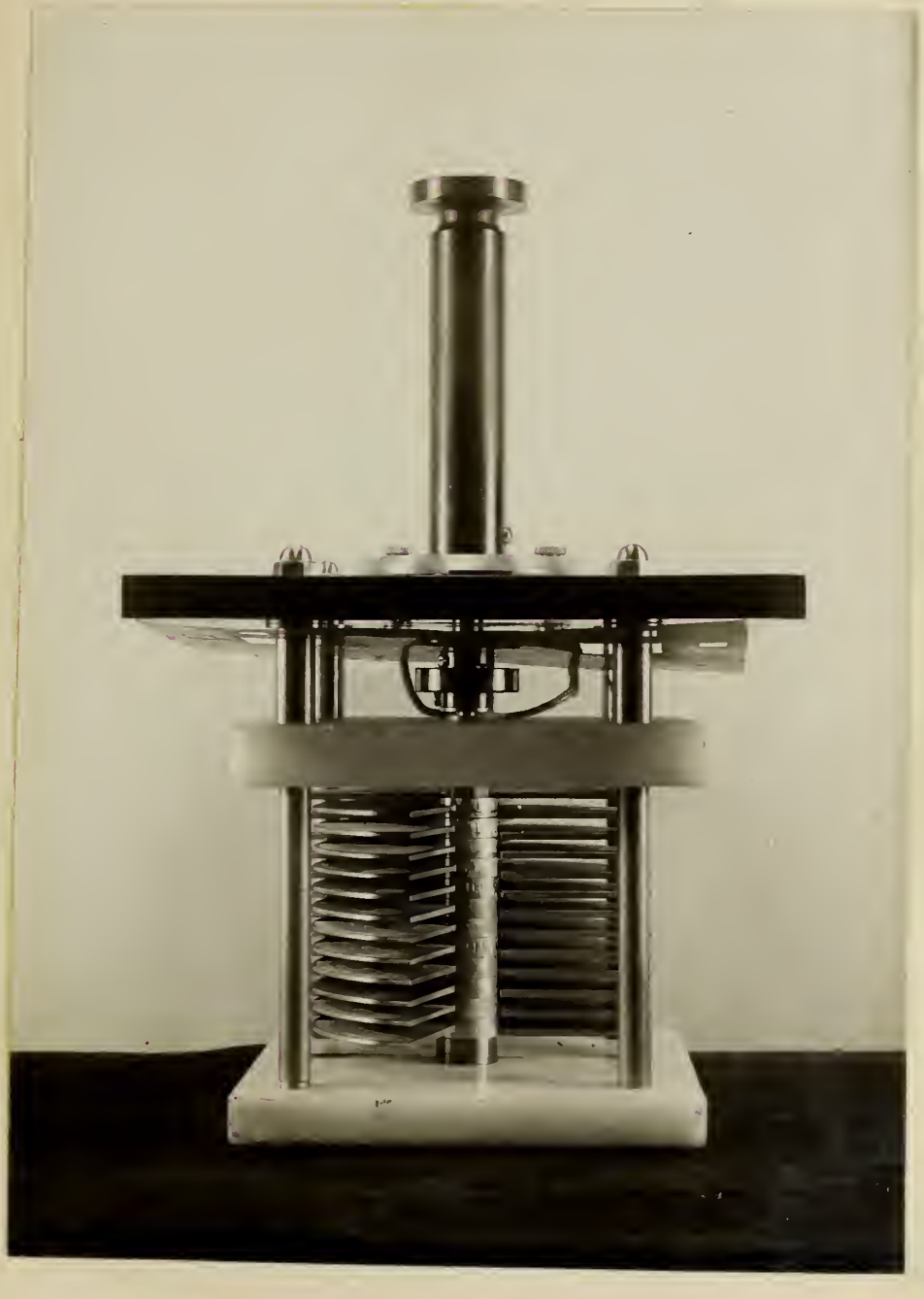


Fig. 21

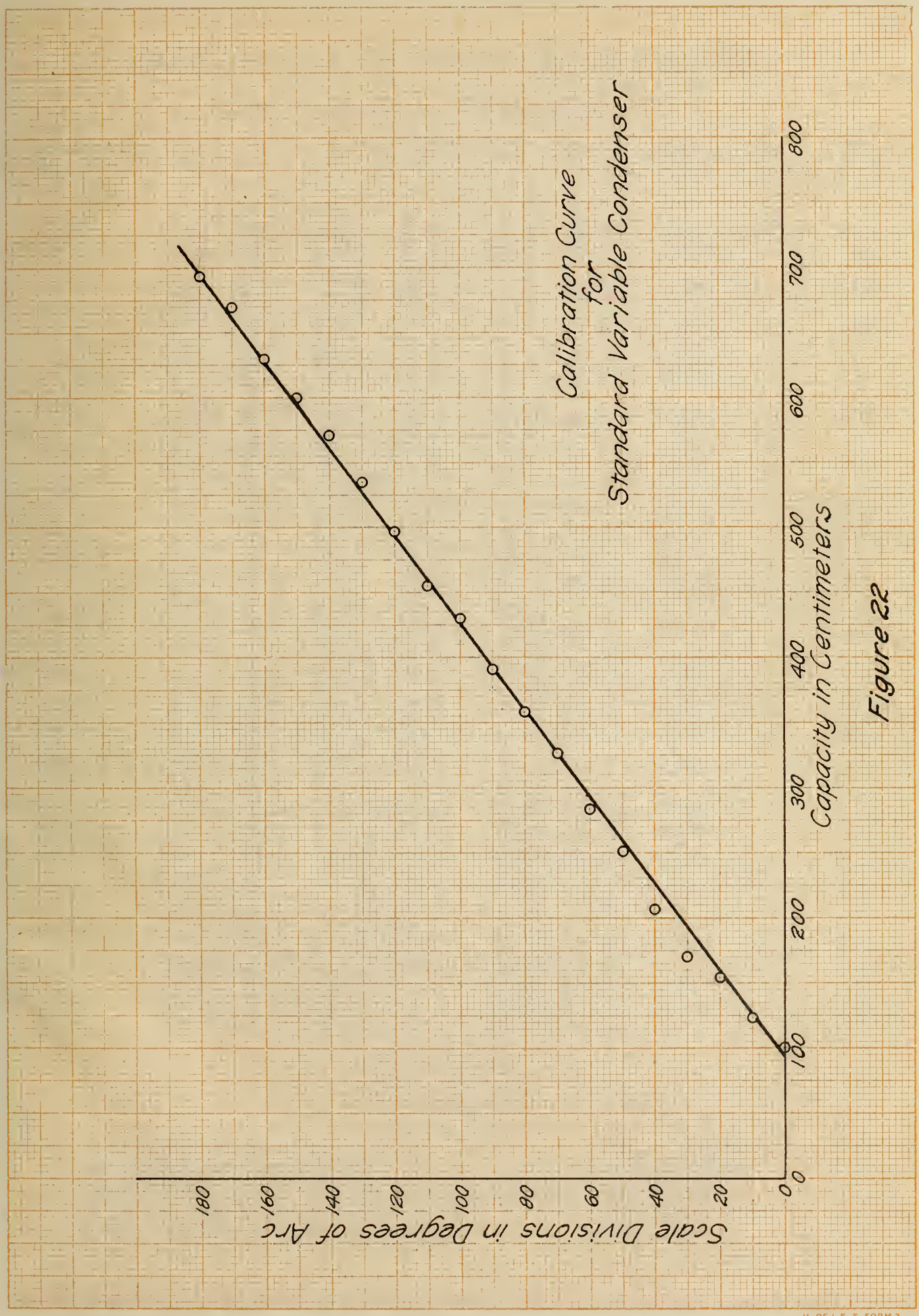


Figure 22

U. OF I. S. S. FORM 1

CALIBRATION DATA FOR STANDARD VARIABLE CONDENSER

taken by Mr. S. F. Tanabe, Feb. 1917

Condenser Scale	d_o	d_1	d_2	D_1	D_2	D_1-D_2	$\frac{D_1-D_2}{D_2}$	C_s	$C_s\frac{D_1-D_2}{D_2}$
0	11.2	42.3	30.1	31.1	18.9	12.2	.645	155.5	100.3
	11.2	42.3	30.2	31.1	19.0	12.1	.637		99.2
10	11.2	42.3	28.5	31.1	17.3	13.7	.791		123.0
	11.1	42.2	28.45	31.1					
20	11.2	42.3	26.6	31.1	15.6	15.5	.994		154.5
	11.1	42.2	26.7	31.1					
30	11.1	42.1	25.9	31.0	14.8	16.2	1.095		170.5
	11.1	42.1	25.95	31.0					
40	11.35	42.35	24.8	31.0	13.45	17.7	1.32		207.0
	11.4	42.4	24.6	31.0	13.2				
50	11.5	42.6	23.4	31.1	11.9	19.15	1.61		251.0
	11.55	42.55	23.45	31.0					
60	11.1	42.1	22.10	31.0	11.0	20.0	1.82		283.0
	11.1	42.1	22.10	31.0					
70	11.1	42.1	21.1	31.0	10.0	21.0	2.10		327.0
80	11.1	42.1	20.6	31.0	9.5				
	11.35	42.4	20.8	31.05	9.4	21.6	2.30		358.0
	11.4	42.4	20.9	31.00	9.3				
90	11.35	42.4	20.3	31.05	8.95	22.2	2.52		391.0
	11.40	42.4	20.15	31.0	8.75				
100	11.40	42.5	19.55	31.1	8.25	22.85	2.77		430.0
	11.40	42.5	19.60	31.1	8.20				
110	11.40	42.5	19.3	31.1	7.90	23.1	2.93		455.0
120	11.40	42.5	18.8	31.1	7.40	23.7	3.20		497.0
	11.40	42.5	18.8	31.1					
130	11.40	42.5	18.4	31.1	7.00	24.1	3.44		535.0
140	11.40	42.5	18.05	31.1	6.65	24.45	3.675		571.0
	11.40	42.5	18.03	31.1	6.63				
150	11.40	42.5	17.80	31.1	6.40	24.70	3.86		600.0
160	11.40	42.5	17.55	31.1	6.15	24.95	4.06		630.0
	11.40	42.5	17.50	31.1	6.10				
170	11.40	42.5	17.25	31.1	5.85	25.25	4.32		670.0
	11.40	42.5	17.25	31.1	5.85				
180	11.40	42.5	17.05	31.1	5.65	25.4	4.46		693.0
	11.35	42.45	17.05	31.1	5.70				
0	11.35	42.45	30.25	31.1	18.90	12.2	.645		100.4
30	11.35	42.45	25.70	31.1	14.35	16.75	1.165		181.5
	11.30	42.40	25.60	31.1	14.30				

$$D_1 = d_1 - d_o$$

$$D_2 = d_2 - d_o$$

Potential on needle	60. volts
Potential on quadrants	2.8 volts
Copy of Standard cylindrical condenser	122.4 cm.
Capacity of electrometer	33.1 cm.
Total comparison capacity C_s	155.5 cm.

standard. All other condensers used were of the ordinary type of variable condenser sold by radio supply firms.

Each of the inductance coils shown as L_2 in the diagrams was 13 centimeters in diameter and 64 centimeters long, and consisted of a single layer of No.28 silk-covered copper wire. Each coil was provided with taps dividing it into ten equal sections. The coils shown as L_1 in the diagrams each consisted of a single layer of No.24 silk-covered copper wire, and were 9 centimeters in diameter and 6.5 centimeters in length. The coils L_3 and L_4 of the primary circuit were ring inductances 25 centimeters in diameter mounted in variometer form. The inductance of each was approximately sixteen millihenries. Two types of vacuum tube were used. In the primary circuit a Western Electric type VT-1 tube was used. The normal filament current was 1.1 amperes. In the test circuit a tube similar to the present "audiotron" type was used. The normal filament current for this tube was 0.7 ampere. The current for each tube was supplied by a large six-volt storage battery in series with a small dial resistance. Dry cells were used for the B batteries, the voltage used being from 22 1/2 to 24 volts.

The telephones used were two Brandes head sets, each of 7000 ohms resistance.

VIII RESULTS

Determinations were made of the dielectric constants of benzene, toluene, carbon tetrachloride, and two samples of petrol ether. The frequency was approximately 75000, and the temperature 24°C. The results are tabulated below:

Benzene

R_o	R_1	R_2	R_1-R_o	R_2-R_o	k
155.50	118.10	70.60	37.40	84.90	2.270
161.60	114.90	56.20	46.70	105.40	2.257
161.90	124.30	77.00	37.60	84.90	2.258
161.80	115.10	56.20	46.70	105.60	2.261
162.90	113.80	51.90	49.10	111.00	2.261
150.80	113.30	65.90	37.50	84.90	2.264
				Mean	2.260

Toluene

R_o	R_1	R_2	R_1-R_o	R_2-R_o	k
163.30	118.40	57.00	44.90	106.30	2.367
163.10	121.50	64.60	41.60	98.50	2.367
163.10	119.40	59.90	43.70	103.20	2.361
150.80	110.70	56.30	40.10	94.50	2.357
150.60	113.40	63.00	37.20	87.60	2.355
151.10	113.90	63.00	37.20	88.10	2.368
				Mean	2.362

Carbon Tetrachloride

R_o	R_1	R_2	R_1-R_o	R_2-R_o	k
166.20	128.90	83.30	37.30	82.90	2.222
166.00	128.30	82.40	37.70	83.60	2.217
157.70	120.30	75.00	37.40	82.70	2.211
157.30	120.00	74.20	37.30	83.10	2.227
155.40	118.00	72.10	37.40	83.30	2.227
				Mean	2.221

Petrol Ether I (B.P. 45-65°C.)

R_o	R_1	R_2	R_1-R_o	R_2-R_o	k
153.80	115.20	79.80	38.60	74.00	1.917
154.80	117.50	83.10	37.30	71.70	1.922
154.80	117.70	83.10	37.10	71.70	1.932
154.70	117.60	82.70	37.10	72.00	1.940
150.20	113.00	78.20	37.20	72.00	1.935
151.10	113.90	79.00	37.20	72.10	1.938
151.10	113.90	78.80	37.20	72.30	1.943
				Mean	1.932

Petrol Ether II (B.P. 30-50°C.)

R_o	R_1	R_2	R_1-R_o	R_2-R_o	k
150.20	113.00	82.00	37.20	68.20	1.833
150.30	112.80	81.70	37.50	68.60	1.829
150.40	113.10	81.90	37.30	68.50	1.836
146.30	109.10	77.80	37.20	68.50	1.841
161.80	119.30	83.40	42.50	78.40	1.845
166.30	126.40	92.40	39.90	73.90	1.852
				Mean	1.839

With the vernier provided on the condenser scale it was estimated that readings could be taken to within 0.035 or 0.04 degree. Then the error in determining the capacity of the condenser with air as the dielectric should be within 0.2 per cent. In the case of a liquid of small dielectric constant, such as sample II of petrol ether, the error should be about 0.1 per cent. Then the error in the value of the dielectric constant should be approximately 0.3 per cent or less.

In the above table of data, the largest variation from the mean value is 0.18 per cent in the case of benzene. For toluene the greatest variation from the mean is 0.3 per cent. The third determination for carbon tetrachloride shows a variation from the mean of 0.45 per cent, the others falling within 0.27 per cent. The variation for the first sample of petrol ether was 0.31 per cent, and for the second sample 0.54 per cent excepting for the last reading which differed from the mean by 0.71 per cent.

As to the accuracy secured in applying the heterodyne principle for adjusting the test circuit, a change of the scale position of seventeen degrees produced a change of one thousand in the beat frequency. A change of one-tenth degree in the condenser setting, then, would produce a beat change of about six per second. Since adjustments could be made for which the acoustic beats between the tuning

fork and the beat note in the telephones did not recur as often as once in five seconds, it is seen that the accuracy of the results depends upon the accuracy of the condenser settings.

IX SUMMARY

1. A resonance method using undamped oscillations has been developed for determining dielectric constants of liquids.

2. The heterodyne principle has been shown to be a sensitive means of detecting changes of capacity.

3. The method is simple and manipulation is easy and rapid.

4. Computations are very simple.

5. The method is accurate to approximately 0.3 per cent.

6. Only a small sample of liquid is necessary, thirty cubic centimeters being needed for a determination.

7. The method was unsuccessful for liquids of appreciable conductivity.

In conclusion, the author wishes to express his thanks to Professor A.P. Carman for the facilities for this investigation, and for his assistance and direction.

Acknowledgment is due Mr. S.F. Tanabe for the calibration data for the standard variable condenser, and Mr. J.B. Hays for his excellent work in constructing the condenser.

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von Lang,V. Wied.Ann. 57, p 430, 1896.
Determined dielectric constants by using a Quincke interference tube for determining the ratio of the wave-lengths in air and in the liquid.

Lecher,E. Wied.Ann. 42, p 142, 1891.
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Marx,E. Wied.Ann. 66, p 411, 1898.
Annalen 12, 3, p 491, 1903.
Used Drude method with wave-lengths of 58, 56, and 4 cm. in air. Tested benzol, water and alcohol.
Used frequency 10^7 to 10^8 to see if the dielectric constant were a function of the frequency. No variation of the order of 1 per cent detectible.

Mazotto,D. Acc dei Lincei (5), 4, I, p 240, 1895.
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constant of SO_2, NH_3 and water, to show that a loss of chemical energy was in reality a physical change in the dielectric energy of the system.

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First resonance method to be used. Determined dielectric constants of a large series of substances for frequency of 10^6.

Vonwiller,O.U. Phil.Mag. 7, p 655, 1904.
Determined the dielectric constant of water near 4°C., using a Lecher system. Found dielectric constant diminished as the temperature increased from 0°C.

Walden,P. Acad. Sci. St.Pet., Bull.4, p 305, 1912.
Used Drude method, wave-length 70 cm. Tested dielectric constants of ammonia salts dissolved in chloroform. Temperature 17°C to 19°C. Error 1 per cent to 5 per cent.

Yule,S. Wied.Ann. 50, p 742, 1893.
Determined the dielectric constant of water at 17°C as 73.8, using a Lecher system.

V I T A

William Henry Hyslop was born at Verona, Illinois, December 15, 1883. His grammar school training was received in the public schools of Verona, and also Bloomington, Illinois. He attended high school at Bloomington and at Streator, Illinois. He finished his preparatory school work at Knox Academy, Galesburg, Illinois. He attended Knox College, 1904-1908, and was graduated with the A. B. degree. He was part time assistant in physics at the University of Illinois, 1909-1911, assistant in physics, 1914-1917, and instructor in physics, January 1919 - June, 1920. He taught physics and physiography in the high school at Decatur, Illinois, 1911-1914. He was head of the division of signalling and radio in the United States Army School of Military Aeronautics, University of Illinois, June, 1917 - December, 1918, holding a commission as first lieutenant in the aeronautics branch of the air service during the last four months. His graduate work was done during the period of his service in the physics department of the University of Illinois.

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